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13. ABSTRACT (Maximum 200 words)  We have established the exact nature of the various scattering processes between carriers and the many-body effects that produce large nonlinearities near the bandedge of direct gap III-V semiconductors. The femtosecond optical nonlinearities near the bandedge of GaAs have been investigated after injection of very hot carriers with 2 eV photons. Instantaneous changes of refractive index and absorption coefficient have been found to be consistent with bandgap renormalization and screening of the Coulomb interactions. The combined effects of bandfilling, bandgap renormalization, plasma screening of the Coulomb interactions, and free carrier absorption leads to an optical response that may be exploited in device applications. At high injected densities, the dynamics of gain near the bandedge has been established. It is controlled by electron-electron and electron-hole scattering, and numbers for these effects have been obtained. The initial scattering of the hot carriers has been investigated by hole burning experiments in intrinsic, p-type and n-type GaAs. In some cases, a very short scattering time (<25 fs) has been observed due to emission of coupled plasmon-phonon modes.				
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**Optics with Semiconductors:  
Ultrafast Physics for Devices**

**FINAL PROGRESS REPORT**

**Philippe M. Fauchet**

**September 5, 1996**

**U.S. ARMY RESEARCH OFFICE**

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## FINAL REPORT

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1. **Femtosecond Refractive and Absorptive Nonlinearities Due to Real Carriers in GaAs**, in OSA Proceedings on Picosecond Electronics and Optoelectronics, volume 9, edited by T.C.L.G. Sollner and J. Shah, Optical Society of America, Washington DC, 1991, pp253-259 (*Note that this paper still acknowledges support from ARO grant DAAL03-87-K-0145*)
2. **Femtosecond Refractive-Index Spectral Hole Burning in Intrinsic and Doped GaAs**, Appl. Phys. Lett. **59**, pp 721-723 (1991)
3. **Femtosecond Gain Dynamics due to Initial Thermalization of Hot Carriers Injected at 2 eV in GaAs**, Phys. Rev. B **44**, pp 6542-6545 (1991)
4. **Femtosecond Gain Dynamics in Thin GaAs Films**, presented at Internat. Conf. Hot Carriers in Semiconductors, Nara, Japan, July 1991; Semicond. Sci. & Technol. **7**, B164-B166 (1992)
5. **Femtosecond Nonlinearities and Hot-Carrier Dynamics in GaAs**, INVITED PAPER presented at the Internat. Symp. on Ultrafast Processes in Spectroscopy, Bayreuth, Germany, October 1991; in *Ultrafast Processes in Spectroscopy 1991*, edited by A. Laubereau and A. Seilmeier, Institute of Physics Conference Series No. 126 (IOP Publishing Ltd., Bristol, England, 1992), pp. 317-324. (Invited)
6. **Femtosecond Dynamics of Hot Carriers in GaAs**, INVITED PAPER presented at the SPIE Conf. on Ultrafast Laser Probe Phenomena in Bulk and Microstructure Semiconductors and High-Temperature Superconductors, Somerset NJ, March 1992; *Ultrafast Laser Probe Phenomena in Bulk and Microstructure Semiconductors and High-Temperature Superconductors*, edited by R. R. Alfano (SPIE, Bellingham, WA, 1992), Vol. 1677, pp. 25-34. (Invited)
7. **Carrier-Carrier Interactions in GaAs Investigated by Femtosecond Spectroscopy**, presented at the SPIE Conf. on Ultrafast Pulse Generation and Spectroscopy, Los Angeles, January 1993; in *Ultrafast Pulse Generation and Spectroscopy*, edited by T.R. Gosnell et al (SPIE, Bellingham, WA 1993), Vol. 1861, pp. 227-236.

- 8. Femtosecond Optical Response of Low Temperature Grown InGaAs**, J. Electron. Mater. **22**, 1477-1480 (1993).
- 9. Subpicosecond Gain Dynamics in GaAs**, presented at the March Meeting of the American Physical Society, Indianapolis. 1992 (Abstract only)
- 10. Femtosecond Spectral Hole Burning in III-V Semiconductors**, ibidem (Abstract only)

## SUMMARY OF RESEARCH FINDINGS

In our work, we have established the exact nature of the various scattering processes between carriers and the many-body effects that produce large nonlinearities near the bandedge of direct gap semiconductors. In this grant, which is the continuation of a 3-year grant from ARO that ran from 1987 to 1990, we have made several important discoveries.

In a first series of experiments, we have investigated the femtosecond optical nonlinearities near the bandedge of GaAs after injection of very hot carriers with 2 eV photons. The experiments have been performed at room temperature on thin films of GaAs attached to a sapphire window using <100 fs pulses from an amplified CPM dye laser. We have found instantaneous changes of refractive index and absorption coefficient, even below the original bandgap, consistent with bandgap renormalization and screening of the Coulomb interactions (Sommerfeld enhancement). The optical nonlinearity continues to evolve on a picosecond time scale, reflecting the cooling of the carrier distribution which leads to bandfilling. A comparison between our data and the predictions of a model developed for equilibrium conditions shows a qualitative agreement but a quantitative disagreement that we attribute to differences in many-body effects between hot and cold carriers. When we probe 120 meV below the original bandedge (950 nm), we find a small but long-lasting induced absorption, produced by free carrier absorption. The free carrier cross-section absorption for an electron-hole pair is  $2.6 \cdot 10^{-17} \text{ cm}^2$  at 950 nm nearly independent of the carrier temperature. At 950 nm, the change of index of refraction is still large because of the influence of bandfilling. For the development of phase modulators, the ratio of the change of index to that of the change of absorption coefficient multiplied by the wavelength should be as large as possible. We have found that selecting wavelengths well below the bandgap, such as 950 nm, makes this ratio larger than unity. In fact, approximately 20 to 50 meV below the original bandgap, the effects of bandfilling, bandgap renormalization, plasma screening of the Coulomb interactions, and free carrier absorption combine to produce a net absorption equal to 0! This result is very promising and future work will establish the usefulness of this effect in devices as it depends strongly on the injected carrier density and the time delay.

An extreme case of optical nonlinearities is optical gain. Gain occurs near the bandedge when there are more electrons in the conduction band than in the valence band (population inversion). If the carriers are injected with 2 eV photons, that is approximately 0.6 eV excess kinetic energy, we expect that gain would take some time to develop. This time will be governed by the cooling rate of the electrons and of the holes. The holes have usually been assumed to cool rapidly to the lattice temperature, since they can lose energy by many different types of hole-phonon collisions. Electrons on the other hand lose energy by emission of LO phonons only and the electron-LO phonon collision time is  $\sim 180$  fs. Thus, electrons with an excess kinetic energy of  $\sim 500$  meV would take more than 2.5 ps to reach the bottom of the conduction band and thus make gain possible. The situation is further worsened by the screening of the electron-LO phonon interaction at high injected electron densities and by the fast ( $\sim 100$  fs) transfer of electrons from the central valley to the satellite valleys, where they are known to remain for  $\sim 2$  ps. In spite of these considerations, we have been able to observe gain in the vicinity of the bandedge after a time as short as 300 fs. The reason for this is two fold: first, efficient electron-electron scattering produces a very hot but thermalized distribution in the central valley before scattering to the satellite valleys can take place; second, once the hottest electrons in that distribution scatter to the satellite valleys, the electrons remaining in the central valley are effectively quite cold. This two-step mechanism produces a relatively cold electron distribution in a time scale of the order of 100 fs. This distribution is sufficient to provide population inversion, once the holes are very cold. In collaboration with a theoretical group at the National Research Council of Ottawa, we have developed a quantitative model that describes these physical phenomena. Two results must be highlighted: first, at high carrier density, electron-electron scattering does indeed redistribute the carriers throughout the central valley on a time scale much shorter than the experimental resolution of less than 100 fs; second, we have obtained the first measurement of the hole temperature as a function of time. It shows that electron-hole collisions heat the hole temperature during the first

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electrical engineering

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300 fs and that once the electrons have cooled down, the hole temperature then reaches the lattice temperature in less than 1 ps. Our conclusion is that electron-hole interactions are much more efficient than commonly assumed and that the hole-lattice interactions indeed bring the hole temperature down to the lattice temperature very quickly.

In a series of distinct experiments, we have investigated the initial scattering of the hot carriers. Here, the probe is tuned around the pump wavelength. We can thus monitor the rate at which the photoinjected carriers leave their initial excited states. Let us emphasize that this rate is not necessarily the rate at which the carriers form a thermalized distribution, as our measurement monitors only the carriers around the initial excited states. We have previously reported our first observation of the refractive-index spectral hole burning. We have now used this powerful tool to compare the initial scattering rates in intrinsic, p-type and n-type GaAs. To our surprise, at low injected carrier densities, the hole burning that is clearly visible with the intrinsic samples remains unchanged in p-type GaAs but becomes barely observable in n-type GaAs. Furthermore, in n-type GaAs, when the injected carrier density exceeds  $7 \times 10^{17} \text{ cm}^{-3}$ , the hole burning reappears, which indicates that the initial scattering time increases with increasing carrier density! The very short scattering time ( $< 25$  fs) for one hot carrier in the presence of a dense cold electron plasma is due to emission of coupled plasmon-phonon modes, as demonstrated by model calculations. In p-type GaAs, these modes are heavily damped and thus not effective. The reason for the increase of the scattering time as the injected carrier density increases will continue to be under active investigation (novel three pulse experiments, samples with different doping densities, experiments at different lattice temperatures); our preliminary results suggest that it is the photoinjected holes (not electrons) which are responsible for the reduced scattering rate.

Finally, our hole burning results at 2 eV in GaAs held at room temperature show that the peak of the hole is shifted from 620 to 650 nm. We speculate that this results from an ultrafast scattering of the electrons injected from the heavy-hole band which brings them to the bottom of the X-valley and then back again to the central valley. This model could explain our result and results obtained with 10 fs pulses by C.V. Shank et al in 1990. It is consistent with the much smaller shift seen when the spectral hole is burned below the L-valley minimum, the absence of a shift we have recorded in InP (a material in which there is no intervalley scattering), and the apparent lack of shift with GaAs at low temperature.